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Absolute Yields of ^{99}Mo in the Thermal Neutron-Induced Fission of ^{233}U and ^{239}Pu

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Summary

Absolute yields of ^{99}Mo were redetermined in the thermal neutron-induced fission of ^{233}U and ^{239}Pu. $(4.75 \pm 0.11)\%$ and $(6.22 \pm 0.14)\%$ were obtained, respectively, for the yields of ^{233}U and ^{239}Pu.

Zusammenfassung

Die absoluten Ausbeuten von ^{99}Mo wurden bei der thermischen Neutronenspaltung von ^{233}U und ^{239}Pu neu bestimmt. Es wurden die folgenden Werte erhalten: ^{233}U: $(4{,}75 \pm 0{,}11)\%$; ^{239}Pu: $(6{,}22 \pm 0{,}14)\%$.

Résumé

La production du ^{99}Mo dans la fission du ^{233}U et du ^{239}Pu par les neutrons thermiques a été redeterminée. Les valeurs suivantes ont été trouvées: ^{233}U: $(4{,}75 \pm 0{,}11)\%$ et ^{239}Pu: $(6{,}22 \pm 0{,}14)\%$.

Molybdenum-99 is an important standard nuclide for the determination of fission yields. Whereas general agreement has been reached for its yield in the thermal neutron fission of ^{235}U [1–7], there exist still large discrepancies between different measurements for ^{233}U and especially for ^{239}Pu [8–15]. The variations in the experimental values are of course reflected in the evaluations of fission yields which recommend different "best" values [1–6]. An attempt for a reliable redetermination of the absolute fission yields of ^{99}Mo in the thermal neutron-induced fission of ^{233}U and ^{239}Pu seemed justified.

Our absolute method was successfully used for the determination of ^{99}Mo and ^{139}Ba in the thermal neutron-induced fission of ^{235}U, and was described in detail in an earlier publication [7]. Sample preparation, irradiation, fission counting and chemical separation for the ^{233}U and ^{239}Pu were performed similar to the measurements for ^{235}U fission. The isolated molybdenum samples were counted on calibrated NaI(Tl)-, Ge(Li)-, and proportional counters. The calibration was performed with two standard solutions of ^{99}Mo from the Radiochemical Centre Amersham (England). Furthermore, the Ge(Li)-detector (resolution 1.95 keV for 1332 keV) was also calibrated with other standard nuclides from the IAEA, Vienna and from the Radiochemical Centre Amersham. For the measurements on the NaI(Tl)-detector the area under the 140 keV photopeak of ^{99m}Tc was calculated after reaching transient equilibrium with ^{99}Mo. For the measurements on the Ge(Li)-detector the gamma-lines at 140.5 keV (^{99m}Tc) and 181.1 keV were used, with intensivities of $(89.6 \pm 0.5)\%$ and $(6.7 \pm 0.2)\%$ [16], respectively. These abundances include corrections for the ^{99}Mo–^{99m}Tc equilibrium and for conversion electrons. Corrections for self absorption were made for the proportional counter. The decay of the molybdenum samples was followed on all three detection systems in order to check for radiochemical purity.

The results of the determinations of the fission yield of ^{99}Mo are shown in Tables 1 and 2. Ten determinations were performed for ^{233}U and nine for ^{239}Pu. The fission rate R_f in the tables includes a correction for counting efficiency and for counting losses. R_f is multiplied by the saturation factor $(1 - e^{-\lambda t})$. A half-life of 66.7 hrs [17] was used for ^{99}Mo. The irradiation times t were of the order of 70 to 80 hrs. The activities in Tables 1 and 2 are corrected for chemical yield, detector efficiencies and decay properties of ^{99}Mo. If the measurements were performed on several detection systems mean values of the different determinations were used to compute the activity of the samples.

The statistical error in the fission counting, including corrections for efficiency and coincidence losses is believed to be much smaller than $\pm 1\%$. The neutron

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Table 1. *Summary of the experiments for the determination of* ^{99}Mo *in the thermal neutron-induced fission of* ^{233}U

Experiment	Fission rate $R_f \cdot (1 - e^{-\lambda t})$ fissions/min	Activity of ^{99}Mo at end of irradiation disintegrations/min				Fission yield %
		Proportional counter	NaI-detector	Ge(Li)-detector		
				140 keV	181 keV	
1	352'822		17'234			4.88
2	188'572		8'994			4.77
3	302'761		14'275			4.71
4	335'955		16'219			4.83
5	209'273		9'603			4.59
6	240'481		11'402			4.74
7	265'657		12'534			4.72
8	365'755	16'585		16'984	15'901	4.51
9	428'873	21'483		19'551	18'812	4.65
10	441'070	22'182		23'055	22'603	5.13
					Average:	$(4.75 \pm 0.11)\%$[a]

[a] Including systematic error of 2%.

flux was stable to $\leq \pm 2\%$ during all the measurements. The errors in the activity measurements are of the order of $\pm 5\%$, this including statistical errors in the β- and γ-counting of the samples ($\leq \pm 1\%$), an estimated error in the detector efficiency calibration of about $\pm 4\%$, estimated errors in the decay schemes ($\pm 2\%$) and errors in the determination of the chemical yield ($\pm 2\%$). As only the fission rate and the activity were determined in our straight-forward method no errors due to measurements of the neutron flux, the amount of fissionable material and uncertainties in the fission cross-sections (variations with the neutron temperature) are introduced. Systematic errors could occur in the calibration of the detectors. However, they are expected to be small, since the results obtained with the different detection systems are in agreement and do not show a systematic deviation. Furthermore, an independent calibration with a set of different standard samples was performed for the Ge(Li)-detector. Systematic errors should cancel out in this calibration. We believe that the main source of systematic errors lies in uncertainties of the decay schemes. The use of additional D_2O-moderators in the neutron beam did not affect the measurements. Therefore, systematic errors due to non-thermality of the neutron spectrum ought to be small.

The errors indicated for the average of the different determinations in Tables 1 and 2 are one standard deviation. They were calculated on the basis of the spread of the individual measurements and on the estimated systematic error in the decay schemes of 2%.

Table 3 shows a comparison of the published values for the fission yield of ^{99}Mo in the fission of ^{233}U and ^{239}Pu. Our result of $(4.75 \pm 0.11)\%$ for ^{233}U is outside of one standard deviation of the mean of all the former

Table 2. *Summary of the experiments for the determination of* ^{99}Mo *in the thermal neutron-induced fission of* ^{239}Pu

Experiment	Fission rate $R_f \cdot (1 - e^{-\lambda t})$ fissions/min	Activity of ^{99}Mo at end of irradiation disintegrations/min			Fission yield %
		Proportional counter	Ge(Li)-detector		
			140 keV	181 keV	
1	88'565		5'539	5'399	6.18
2	71'570		4'335		6.06
3	325'545	20'988	22'039	20'311	6.49
4	325'952	20'610	20'483	19'170	6.16
5	173'212	11'617	11'218	8'912	6.11
6	227'563	15'711	15'568	14'300	6.68
7	405'688	24'465	24'494	24'934	6.07
8	461'015	28'080	27'884	26'760	5.98
9	248'882	15'730	15'597	15'628	6.29
				Average: $(6.22 \pm 0.14)\%$[a]	

[a] Including systematic error of 2%.

Table 3. *Published values for the yields of* ^{99}Mo *in the thermal neutron-induced fission of* ^{233}U *and* ^{239}Pu

Authors		% Fission yield of ^{99}Mo ^{233}U	^{239}Pu
a) *Measurements*			
Steinberg and Glendenin	[8]	5.1	6.1
Ford and Gilmore	[9]	4.96 ± 0.15	6.02 ± 0.18
Marsden and Yaffe	[10]		5.61 ± 0.33
Fickel and Tomlinson	[11]		6.44[a]
Kirby	[12]		6.10
Lisman *et al.*	[13]	5.16 ± 0.07	6.59 ± 0.18
Sorokina *et al.*	[14]		6.17 ± 0.19
Jain and Ramaniah	[15]		6.82 ± 0.15
This work		4.75 ± 0.11	6.22 ± 0.14
b) *Evaluations*			
Katcoff	[1]	4.80	6.10
Croall	[2]	4.96 ± 0.25	5.86 ± 0.18
Crouch	[3]	5.08 ± 0.15	6.20 ± 0.19
Meek and Rider	[4]	5.02 ± 0.10	6.43 ± 0.13
Lammer and Eder	[5]	4.89	6.33
Walker	[6]	4.99 ± 0.10	6.32 ± 0.20
Recommended value this work		4.90 ± 0.08	6.23 ± 0.13

[a] Interpolated value.

experimental determinations (5.07 ± 0.06)%. A value of 4.80%, which is often found in the literature was not used for this average, because it was already updated in the paper of STEINBERG and GLENDENIN [8]. Our value for ^{239}Pu of (6.22 ± 0.14)% is in perfect agreement with the mean of the former experimental results of (6.23 ± 0.13)%.

The recent publication of JAIN and RAMANIAH [15] gives a value of 6.82% for the yield of ^{99}Mo in the fission of ^{239}Pu. This result was mainly based on a determination of ^{99}Mo relative to the yield in the fission of ^{235}U. LAMMER and EDER [5] have recalculated the result of JAIN and RAMANIAH, taking into account the influence of different neutron temperatures on the cross-sections used and show that the value of 6.82% can be in considerable error. Based on the mean of the earlier measurements and on our new experimental results we recommend values of (4.90 ± 0.08)% and (6.23 ± 0.13)% for the yield of ^{99}Mo in the thermal neutron-induced fission of ^{233}U and ^{239}Pu, respectively.

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